



# The origin of chondrules: Constraints from matrix composition and matrix-chondrule complementarity



Herbert Palme<sup>a,\*</sup>, Dominik C. Hezel<sup>b,c</sup>, Denton S. Ebel<sup>d</sup>

<sup>a</sup> Forschungsinstitut und Naturmuseum Senckenberg, Senckenberganlage 25, D-60325 Frankfurt am Main, Germany

<sup>b</sup> Institut für Geologie und Mineralogie, Universität zu Köln, Zùlpicherstrasse 55, Germany

<sup>c</sup> Natural History Museum, Department of Mineralogy, Cromwell Road, SW7 5BD, London, UK

<sup>d</sup> Department of Earth and Planetary Sciences, American Museum of Natural History, Central Park West at 79th Street, New York, NY 10024-5192, USA

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## ABSTRACT

One of the major unresolved problems in cosmochemistry is the origin of chondrules, once molten, spherical silicate droplets with diameters of 0.2 to 2 mm. Chondrules are an essential component of primitive meteorites and perhaps of all early solar system materials including the terrestrial planets. Numerous hypotheses have been proposed for their origin. Many carbonaceous chondrites are composed of about equal amounts of chondrules and fine-grained matrix. Recent data confirm that matrix in carbonaceous chondrites has high Si/Mg and Fe/Mg ratios when compared to bulk carbonaceous chondrites with solar abundance ratios. Chondrules have the opposite signature, low Si/Mg and Fe/Mg ratios. In some carbonaceous chondrites chondrules have low Al/Ti ratios, matrix has the opposite signature and the bulk is chondritic. It is shown in detail that these complementary relationships cannot have evolved on the parent asteroid(s) of carbonaceous chondrites. They reflect preaccretionary processes. Both chondrules and matrix must have formed from a single, solar-like reservoir. Consequences of complementarity for chondrule formation models are discussed. An independent origin and/or random mixing of chondrules and matrix can be excluded. Hence, complementarity is a strong constraint for all astrophysical–cosmochemical models of chondrule formation. Although chondrules and matrix formed from a single reservoir, the chondrule–matrix system was open to the addition of oxygen and other gaseous components.

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## 1. Introduction

Chondritic meteorites sample primitive solar system material. Bulk chondrites have not been modified by melting and crystallization processes. Their major element compositions come close to the composition of the Sun (e.g., Lodders et al., 2009). Chondrules are a major structural component of chondritic meteorites. They have experienced temperatures of up to 2000 K with subsequent rapid cooling (Hewins, 1997). The process responsible for making chondrules remains unknown. Suggestions for their formation encompass a wide range of possible mechanisms, such as condensation from a hot solar gas, formation near the Sun and transport with protostellar jets to the asteroidal belt and beyond, collisions of molten planetesimals, heating by shock waves, generated by gravitational instabilities in the solar nebula or by supersonic planetesimals, or heating by electromagnetic processes

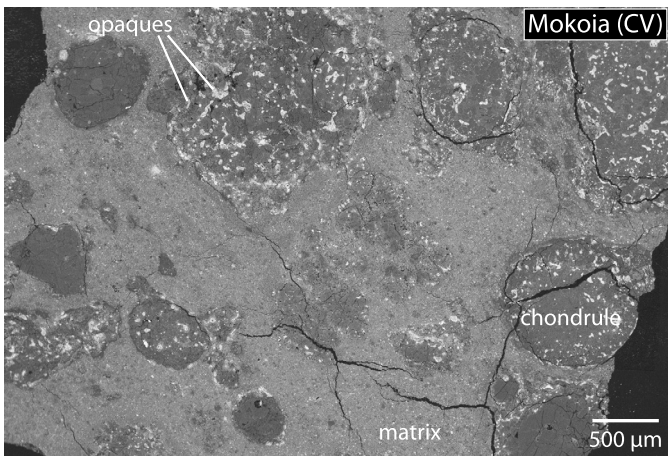
(Ciesla, 2005). A wealth of chemical and isotopic data on single chondrules in a large variety of chondritic meteorites has been collected during the last 40 years (e.g., Jones et al., 2005). Progress has been made in identifying time and conditions of formation of chondrules. The chondrule formation process is arguably the major unresolved question in cosmochemistry.

Carbonaceous chondrites, a sub-group of chondritic meteorites, contain in addition to chondrules large fractions of matrix, from almost 100% in CI-chondrites to 40% or less in the Renazzo type CR-chondrites (e.g., Fig. 1). Many carbonaceous chondrites (e.g., CO, CV) have about equal amounts of chondrules and fine-grained matrix. Matrix is generally crystalline with grain sizes typically below 5 μm (Weisberg et al., 2006). In some of the very primitive CR-chondrites matrix is largely amorphous, i.e. non-crystalline (Abreu and Brearley, 2010).

The bulk chemistry of carbonaceous chondrites is defined by the two major components, chondrules and matrix. Although Ca,Al-rich inclusions (CAI) are an important component in carbonaceous chondrites, their abundance is often overestimated

\* Corresponding author.

E-mail address: [Palmeherbert@gmail.com](mailto:Palmeherbert@gmail.com) (H. Palme).



**Fig. 1.** Chondrules and matrix in the Mokoia CV-chondrite imaged with back-scattered electrons. The darker chondrules indicate lower FeO contents. Bright spots are sulfides or metal.

(Hezel et al., 2008). We estimate a maximum contribution of CAIs to bulk meteorite Si of about 2% to 3% and even less for Mg.

Most chondrules in unmetamorphosed carbonaceous chondrites are dominated by FeO-poor olivine (type 1 chondrules). They are particularly prominent in CR-chondrites. The low Si/Mg ratios of chondrules in these meteorites are compensated by high Si/Mg ratios in matrix, leading to chondritic or solar Si/Mg ratios of the bulk meteorites. This complementarity has been demonstrated for the CR-chondrite Renazzo by Klerner (2001), Klerner and Palme (1999) and for other types of carbonaceous chondrites (Hezel and Palme, 2008, 2010; Ebel et al., 2009). Earlier, Wood (1963, 1985) had elaborated on similar relationships based on differences of Fe/Mg in chondrules and matrix of CM-chondrites.

In this paper we focus on the composition of the matrix in carbonaceous chondrites. Since the submission of two earlier publications on this topic by Hezel and Palme (2008, 2010) several papers with precise and careful measurements of matrices in carbonaceous chondrites have appeared in the literature (Wasson and Rubin, 2009, 2010a; Abreu and Brearley, 2010). These studies showed that matrix is compositionally rather uniform and has a distinct chemical signature, characteristic of all carbonaceous chondrites analyzed. In addition, Stracke et al. (2012) have shown that the relatively coarse grained Allende meteorite is compositionally uniform with regard to Si, Mg and Fe on a scale of a few millimeters. This is significant, because Si, Mg, and Fe plus oxygen make up nearly 90% of the Allende meteorite. Other carbonaceous chondrites should at least be similarly homogeneous. This is also implicitly clear from the generally good agreement of chemical analyses of carbonaceous chondrites by various authors, as discussed below. Here we combine so far scattered data sets to demonstrate the chemical complementarity between chondrules and matrix and discuss possible reasons for it.

## 2. Materials and methods

In this paper we use data on bulk compositions, chondrules and matrix of carbonaceous chondrites. In addition to literature data we report new matrix data of the CV-meteorite Mokoia to complement the data of nearly 100 individual chondrules in Mokoia by Jones and Schilk (2009). Matrix areas from a thin section of Mokoia from the Natural History Museum, London were analyzed with a Cameca SX100 electron microprobe. The accelerating voltage was set to 20 kV, the beam current to 20 nA. The electron beam was defocused to a diameter of 20  $\mu\text{m}$ . The following standards have been used (detection limits in wt.%): Si, synthetic fayalite: 0.02; Ti, synthetic rutile: 0.04; Al, synthetic corundum: 0.02; Cr, synthetic

chromium oxide: 0.04; Fe synthetic fayalite: 0.04; Mn synthetic MnTiO<sub>3</sub>: 0.06; Ni synthetic NiO: 0.06; Mg synthetic forsterite: 0.02; Ca, natural wollastonite: 0.03; Na, jadeite: 0.04. The built in PAP-algorithm (e.g., Pouchou and Pichoir, 1991) was used for correction. Typical totals are around 90 wt.%. This is either due to the occurrence of some hydrous minerals and/or a result of the porous nature of the Mokoia matrix. In the latter case absolute element concentrations might be underestimated by up to 10%. Element ratios are unaffected. Iron in metal and sulfide is calculated as FeO, which leads to artificially high FeO concentrations. Hezel et al. (2013) report a metal + sulfide abundance of 3.3 vol.%, and Ebel et al. (2009) only 0.8 area% for Mokoia. A significant contribution to these values are large metal/sulfide grains, that were avoided during our matrix measurements. As matrix in Mokoia typically has FeO concentrations of around 35 wt.%, and most opaques phases are sulfide (Hezel et al., 2013), the overestimation of matrix FeO from opaque phases is negligible. The Mokoia matrix data are given in the Supplementary Online Material.

We also discuss the chondrule-matrix relationship in Renazzo. Here we are using recent matrix data from Hezel and Palme (2010) and chondrule data from Klerner (2001). The latter data and analytical details are given in the Supplementary Online Material.

## 3. Bulk compositions of carbonaceous chondrites are well defined and approximately solar

Wiik (1956) pointed out the compositional uniformity of bulk carbonaceous chondrites. The average carbonaceous chondrite Si/Mg weight ratio of 22 samples analyzed by Wiik (1956) using wet chemical procedures is  $1.102 \pm 0.034$  (see compilation by Ahrens, 1965). Later independent wet chemical analyses by Jarosewich (1990) gave  $1.109 \pm 0.036$  (19 samples). An average carbonaceous chondrite Si/Mg ratio of  $1.102 \pm 0.025$  (22 samples) was determined by Wolf and Palme (2001) using X-ray fluorescence (XRF). In these data sets there is a small, barely resolvable decrease in the Si/Mg ratio of 3 to 4% in the sequence CI, CM, CV chondrites. Allende (CV) bulk samples have, for example, an average Si/Mg ratio of  $1.080 \pm 0.061$  (Stracke et al., 2012) slightly below the CI-ratio of  $1.12 \pm 0.05$  (Lodders et al., 2009), in accord with a Si/Mg ratio of  $1.080 \pm 0.011$  for an average of a 4 kg Allende sample (Jarosewich et al., 1987). The solar photospheric Si/Mg ratio of  $1.10 \pm 0.2$  fits with the carbonaceous chondrite ratios, but has larger uncertainties (Lodders et al., 2009).

In Fig. 2 we plot the Si/Mg weight ratios of 39 bulk Allende samples, each with a mass of about 600 mg (Stracke et al., 2012). There is a spread of about 5% within the bulk samples, reflecting analytical uncertainties. Only a single sample containing a large Ca, Al-rich inclusion has a different Si/Mg ratio. Also shown are bulk carbonaceous chondrite Si/Mg ratios (Wolf and Palme, 2001). For CR-chondrites the data from Wiik (1956) for Renazzo and Al Rais were taken, in addition to two XRF analyses for Renazzo from Klerner (2001). The Si/Mg ratios in CR-chondrites range from 1.10 to 1.21, in accord with other carbonaceous chondrites.

Fe/Mg ratios among carbonaceous chondrites are somewhat less constant than Si/Mg ratios. The mean Fe/Mg ratios (by mass) decrease from CI (1.92) through CM (1.77) and CO (1.74) to CV (1.55), according to Wolf and Palme (2001). Kallemeyn and Wasson (1981) report a similar decrease: 1.88 (CI), 1.79 (CM), 1.71 (CO) and 1.64 (CV). The Fe/Mg ratios in the 600 mg Allende samples are, however, as constant as the Si/Mg ratios (Stracke et al., 2012). Thus ratios among the three elements Si, Mg and Fe are constant within a few percent in a single carbonaceous chondrite at a 600 mg sample scale, as demonstrated for Allende, a comparatively coarse grained rock.

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